#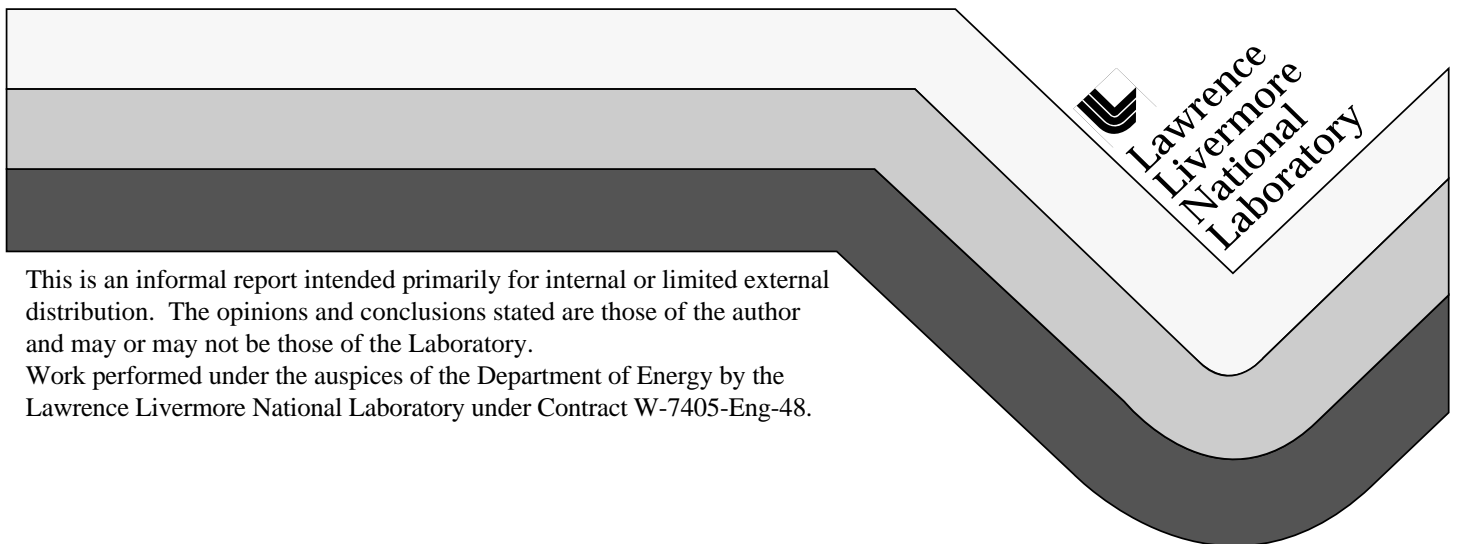 An Analytical Electron Distribution Function for Inelastic Collisions in a Uniform Gas with Time Varying Electric Field

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21 July 1994

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An Analytical Electron Distribution Function for Inelastic Collisions in a Uniform Gas with Time Varying Electric Field

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ABSTRACT

This analysis yields an approximate solution for the isotropic leading term of the spherical harmonic expansion of the Boltzmann equation in the case of a slightly ionized, spatially uniform gas with time varying electric field, and in which inelastic collisions occur. The solution is considered valid for the characteristic frequency of the electric field, defined here as $\omega = (^3/_2)\,(^d/_{dt})\ln[|E_{(t)}| + {}^{kT}/_{e\lambda}]$, less than the electron-molecule elastic collision frequency ν_m. There are no other limiting assumptions made about gas mixture composition, cross-section shapes, or electric field time behavior. This solution is mathematically well behaved for any ratio ω/ν_m. Example distribution functions for both ramp and sinusoidal electric fields are presented for an idealized N_2-like gas.

1

Introduction

      The desire for improved control over electric discharge phenomena in a wide variety of scientific, technological, manufacturing, and waste processing activities spurs the development of non-equilibrium, non-uniform and time dependent models. This paper addresses the situation of a slightly ionized, spatially uniform gas with a time varying electric field, and in which inelastic collisions occur. The purpose here is to present a reasonably consistent, and reasonably accessible analytical result for the electron kinetics in a gas discharge regime of technological interest.

      This paper will be structured as follows. First, the analytical result for the time dependent electron distribution function is stated. Second, a summary of the solution procedure with its attendant assumptions is given. Lastly, examples of the solution are given for an idealized nitrogen-like gas where the electric field ramps between static conditions, and then for sinusoidal behavior.

Analytical Result

      The time dependent distribution function of electron energies is denoted as $f(\varepsilon,t)$, where electron kinetic energy ε is expressed in units of eV, and where the distribution function is normalized as:

$$\int f(\varepsilon,t) \ \sqrt{\varepsilon} \ d\varepsilon = 1, \qquad\qquad (1).$$

The function $B(\varepsilon,t)$ is defined as:

$$B(\varepsilon,t) = -(\partial/\partial\varepsilon) \ \ln[f(\varepsilon,t)], \qquad\qquad (2),$$

and the distribution function is then given by:

$$f(\varepsilon,t) = \frac{e^{-\int_0^{\varepsilon} B(\varepsilon,t)\ d\varepsilon}}{\int_0^{\infty} e^{-\int_0^{\varepsilon} B(\varepsilon,t)\ d\varepsilon} \cdot \sqrt{\varepsilon}\ d\varepsilon}$$

(3).

The logarithmic slope $B(\varepsilon,t)$ is given by:

$$B(\varepsilon,t) = \frac{\sqrt{\left[\mu + \dfrac{\omega}{2\nu_m}\right]^2 + \left[\dfrac{4 \cdot Q_i}{\varepsilon \cdot Q_m}\right] \cdot \left[b + \mu \cdot \dfrac{kT}{e}\right]} + \left[\mu + \dfrac{\omega}{2\nu_m}\right]}{2 \cdot \left[b + \mu \cdot \dfrac{kT}{e}\right]}$$

(4).

The terms used in function $B(\varepsilon,t)$ are as follows:

$$b(\varepsilon,t) = [E/N]^2 / (3\ \varepsilon\ Q_m^2),$$ (5),

$$\omega(t) = \frac{3}{2} * \frac{1}{[E/N]} * \left(\frac{d}{dt}\right) [E/N],$$ (6),

$$\nu_m(\varepsilon) = N * Q_m * \sqrt{2e\varepsilon/m},$$ (7),

$Q_m(\varepsilon)$ = electron-molecule momentum transfer cross section,

$Q_i(\epsilon)$ = electron-molecule inelastic cross section (cm^2),   (8),

$\mu = 2m/M$,

               m = electron mass,

               M = molecule mass,   (9),

kT/e = gas temperature in eV,   (10),

$E(t)$ = time dependent electric field (V/cm) in the form:

$$E(t) = |E(t)| + kT/e * Q_m(kT/e) * N,$$

      $E(t)$ = any imposed field variation over time,   (11),

N = gas mixture number density (cm^{-3}), a constant,   (12).

For a gas mixture:

    δ_s = fractional concentration of species s,

$$Q_m(\epsilon) = \sum_s \delta_s \cdot Q_{m\,s}(\epsilon) \; ,$$

        m = momentum transfer,

$$Q_i(\epsilon) = \sum_s \sum_i \sum_j \delta_{s\,i} \cdot Q_{i\,s\,ij}(\epsilon) \; ,$$

        i = inelastic processes with transition indices:
                i = index of initial energy level,
                j = index of final energy level,

$$\sum_{s}\sum_{i} \delta_{si} = 1$$

,

$$M = \sum_{s} \delta_s \cdot M_s$$

, (13).

Analytical Procedure

This analysis begins with a two term expansion of the electron velocity distribution function in a Boltzmann equation with three collisional effects: recoil and thermal agitation during elastic electron-molecule encounters, and inelastic collisions.[1] The leading term in the expansion describes the bulk heating of the electrons by the electric field in an environment dominated by collisions, while the first order term describes the net drift of this electronic swarm along the field and represents the macroscopic current.

The recoil and thermal agitation collision terms transmit a small fraction of an electron's energy to a molecule, this fraction being given by the ratio of the electron to molecule masses. The inelastic term involves the transfer of sufficient energy to initiate rotational, vibrational, electronic, dissociation, and ionization phenomena. Analytical work on time dependent inelastic electron kinetics goes back at least half a century.[2,3,4,5]

The spatially uniform zeroth and first order velocity distribution equations are transformed to a dependence on electron kinetic energy in units of eV. During this change of parameters the relative velocity between electrons and molecules is ascribed entirely to electron speed. Specifically:

$$\varepsilon = mv^2/2e,$$

$$f_0(\varepsilon,t) = [4\pi\sqrt{2}/(m/e)^{3/2}] * f_0(v,t), \qquad (14).$$

The two equations are combined into a single one for f_0, hence that subscript is eventually dropped. This equation is divided by N

and integrated over energy from 0 to ε. An inelastic cross section is zero below a threshold energy which is at least as large as the energy extracted from the electron during the encounter. For example the cross section for excitation from the 0 to 1 vibrational level in N_2 is zero below about 1.4 eV, while the energy transferred is about 0.3 eV. This fact about the cross sections proves useful in the manipulation of the inelastic collision integrals. The resulting equation is:

$$\sqrt{\frac{m}{2 \cdot e}} \cdot \frac{1}{N} \cdot \frac{\partial}{\partial t} \cdot \int_0^\varepsilon \sqrt{\zeta} \cdot f_0(\zeta, t) \; d\zeta \quad \ldots$$

$$+ \; -\left[\frac{E}{N}\right]^2 \cdot \frac{\varepsilon}{3 \cdot Q_m} \cdot \nu_m \cdot e^{-\nu_m t} \cdot \int_0^t e^{\nu_m \tau} \cdot \frac{\partial f_0(\varepsilon, \tau)}{\partial \varepsilon} \; d\tau \quad =$$

$$\sum_s \sum_i \sum_j \delta_{si} \cdot \int_\varepsilon^{\varepsilon + \varepsilon_{sij}} \zeta \cdot f_0(\zeta, t) \cdot Q_{sij}(\zeta) \; d\zeta \quad \ldots$$

$$+ \left[2 \cdot \frac{m}{M}\right]^2 \varepsilon^2 \cdot Q_m \cdot \left[f_0(\varepsilon, t) + \frac{kT}{e} \cdot \frac{\partial f_0}{\partial \varepsilon}\right] \quad \ldots$$

$$+ \; \frac{1}{3} \cdot \frac{E}{N} \cdot \varepsilon \cdot e^{-\nu_m t} \cdot f_1(\varepsilon, 0)$$

$$(15).$$

The relationship between f_0 and f_1 is:

$$\frac{\partial f_1(\varepsilon, t)}{\partial t} + \nu_m(\varepsilon) \cdot f_1(\varepsilon, t) = -\frac{E}{N} \cdot \frac{\nu_m}{Q_m} \cdot \frac{\partial f_0(\varepsilon, t)}{\partial \varepsilon} \qquad (16).$$

The essence of the solution procedure employed here is to make the exponential transform from $f(\varepsilon,t)$ to $B(\varepsilon,t)$, and then to argue as in the WKB method that the equation for $B(\varepsilon,t)$ has a stationary solution.[6] B is like a local inverse electron temperature and its rate of change is assumed to be slower than the elastic collision frequency, specifically:

$$\frac{\varepsilon}{2 \cdot \nu_m} \cdot \frac{\partial B}{\partial t} \longrightarrow 0 \qquad (17).$$

The resulting equation for B includes the effect of the temporal variation of the normalization integral of the distribution function, which is how the temporal variation of the electric field exerts its influence.

The mechanics of deriving the equation for $B(\varepsilon,t)$ involve: substituting equation (3) into equations (15) and (16) in place of f_0, and then dividing equation (15) by f_0 as defined by equation (3). A convenient label for the normalization integral is $C(t)$. The result from equation (15) is:

$$-\sqrt{\frac{m}{2 \cdot e}} \cdot \frac{1}{N} \int_{0}^{\varepsilon} \left[\sqrt{\zeta} \cdot e^{\int_{\zeta}^{\varepsilon} B(\eta, t) \, d\eta} \cdot \frac{\partial}{\partial t} \cdot \left[\int_{0}^{\zeta} B(\eta, t) \, d\eta + \ln[C(t)] \right] \ldots \right] d\zeta \quad \ldots$$

$$+ \left[\frac{E}{N} \right]^{2} \cdot \frac{\varepsilon \cdot \nu_{m}}{3 \cdot Q_{m}} \cdot \int_{0}^{t} \frac{C(t)}{C(\tau)} \cdot e^{\left[\int_{0}^{\varepsilon} [B(\eta, t) - B(\eta, \tau)] \, d\eta \ldots + -\nu_{m} \cdot (t - \tau) \right]} \cdot B(\varepsilon, \tau) \, d\tau \quad =$$

$$= \sum_{s} \sum_{i} \sum_{j} \delta_{si} \cdot \int_{\varepsilon}^{\varepsilon + \varepsilon_{sij}} \zeta \cdot e^{\int_{\zeta}^{\varepsilon} B(\eta, t) \, d\eta} Q_{sij}(\zeta) \, d\zeta \quad \ldots$$

$$+ \left[2 \cdot \frac{m}{M} \right]^{2} \varepsilon^{2} \cdot Q_{m} \cdot \left[1 - \frac{kT}{e} \cdot B(\varepsilon, t) \right] \ldots$$

$$+ \frac{1}{3} \cdot \frac{E}{N} \cdot \varepsilon \cdot e^{-\nu_{m} t} \cdot f_{1}(\varepsilon, 0) \cdot C(t) \cdot e^{\int_{0}^{\varepsilon} B(\eta, t) \, d\eta}$$

(18).

The analysis of this equation now proceeds by a sequence of approximations. The basic assumptions are:

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    i) B is a weak function of ε,
    ii) B is invariant during a collision time,
    iii) C(t) can be approximated as:

$$C(t) \approx \int_0^\infty e^{-\left\langle\sqrt{\frac{3 \cdot Q_m \cdot Q_i}{E(t)/N}}\right\rangle \varepsilon} \cdot \sqrt{\varepsilon} \; d\varepsilon ,$$

(19).

The approximation used for C(t) is based an energy averaged B(ε) found in an earlier analysis of equation (18) in steady state and for m/M taken as zero.[7] Assumptions (19) lead to a sequence of manipulations:

    a) $\int B \, d\varepsilon$ is approximated as $(B^*\varepsilon)$,

    b) the term in equation (18) which contains time derivatives is expressed as two integrals in the form of incomplete gamma functions, and each in turn is approximated by a two point trapezoid rule,

    c) the time integral term of equation (18) is simplified to:

$$B * \int e^{-v_m*(t-\tau)} \, d\tau,$$

as suggested by the condition expressed as equation (17), also the limit of this integral at t = 0 identically cancels the f_1 term in equation (18),

    d) from (19):

$$\partial \ln[C(t)]/\partial t = (^3/_2) \, \partial \ln[\mathbf{E(t)/N}]/\partial t,$$

    e) the inelastic collision integrals in equation (18) are approximated by $\varepsilon * Q_i/B(\varepsilon,t)$, as was done in an earlier analysis.[7] The resulting equation for B(ε,t) is:

$$-\sqrt{\frac{m}{2 \cdot e}} \cdot \frac{\varepsilon^{\frac{5}{2}}}{2 \cdot N} \cdot \frac{\partial B(\varepsilon, t)}{\partial t} \quad - \sqrt{\frac{m}{2 \cdot e}} \cdot \frac{\varepsilon^{\frac{3}{2}}}{2 \cdot N} \cdot \frac{3}{2} \frac{d}{dt} \ln\left[\frac{E}{N}\right] \ldots$$

$$+ \left[\frac{E}{N}\right]^2 \cdot \frac{\varepsilon}{3 \cdot Q_m} \cdot B(\varepsilon, t) \ =$$

$$\frac{\varepsilon \cdot Q_i}{B(\varepsilon, t)} \ + \ \left[2 \cdot \frac{m}{M}\right] \varepsilon^2 \cdot Q_m \cdot \left[1 \ - \ \frac{kT}{e} \cdot B(\varepsilon, t)\right] \tag{20}.$$

This is rearranged to:

$$\frac{\varepsilon}{2 \cdot v_m} \cdot \frac{\partial B(\varepsilon, t)}{\partial t} \ = \ \left[\frac{\left[\frac{E}{N}\right]^2}{3 \cdot \varepsilon \cdot Q_m^2} \ + \ 2 \cdot \frac{m}{M} \frac{kT}{e}\right] \cdot B(\varepsilon, t) \ \ldots$$

$$- \ \frac{Q_i}{Q_m \cdot \varepsilon} \cdot \frac{1}{B(\varepsilon, t)} \ - \ \left[2 \cdot \frac{m}{M} + \frac{\omega(t)}{2 \, v_m}\right],$$

$$\tag{21}.$$

Equation (4) results from applying condition (17) to equation (21).

Limiting cases of $B(\varepsilon, t)$ are:

a) $\omega \to +\infty$, $B \to +\infty$, (infinitely steep f),

b) $\omega \to -\infty$, $B \to 0$, (flat f),

c) $\omega = 0$, DC: (table below)

$$\mu=0 \quad Q_i=0 \quad E/N=0$$

$$B = \frac{\sqrt{3 \cdot Q_m \cdot Q_i}}{E/N}, \quad \text{massless,} \qquad \surd$$

$$B = \frac{\mu}{b + \mu \cdot \dfrac{kT}{e}}, \quad \text{elastic,} \qquad \surd$$

$$B = \frac{1 + \sqrt{1 + \dfrac{4 \cdot Q_i}{e \cdot Q_m} \cdot \dfrac{1}{\mu} \cdot \dfrac{kT}{e}}}{2 \cdot \dfrac{kT}{e}}, \quad \text{thermal,} \qquad \surd$$

$$B = \frac{e}{kT}, \quad \text{elastic \& thermal,} \qquad \surd \qquad \surd.$$

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Examples in a Model Gas

Examples are shown for a model gas with molecular weight $Z = 29$ (like air), a constant $Q_m = 10^{-15}$ cm^2, at temperature $T = 300°$ K, and at number density $N = 3.54 * 10^{16}$ cm^{-3} (1 Torr). An inelastic cross section similar to vibrational excitation in N_2 and with a peak of $Q_i(1.7) = 3 * 10^{16}$ cm^2 was chosen and is shown below.
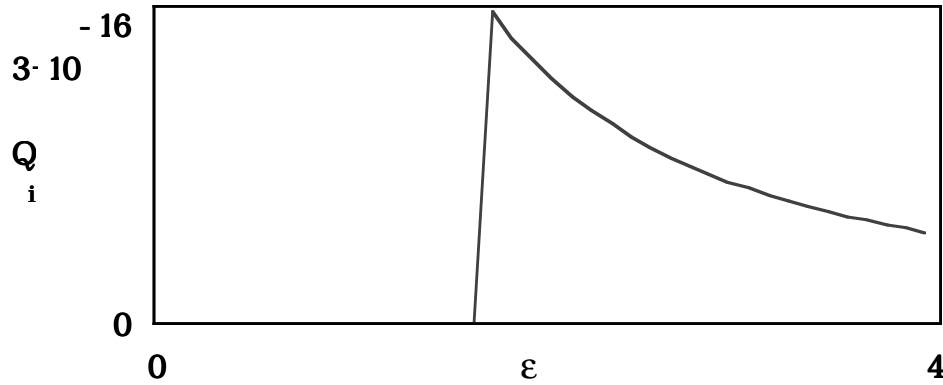
Figure 1: Model Inelastic Cross Section

The first three examples are for an E/N ramps up to 100 Townsends (1 Td = 10^{-17} Volts-cm^2), particulars are noted by each figure (Figures 2 through 5).

The second three examples are for field collapse from an initially steady 100 Td (Figures 6 through 12).
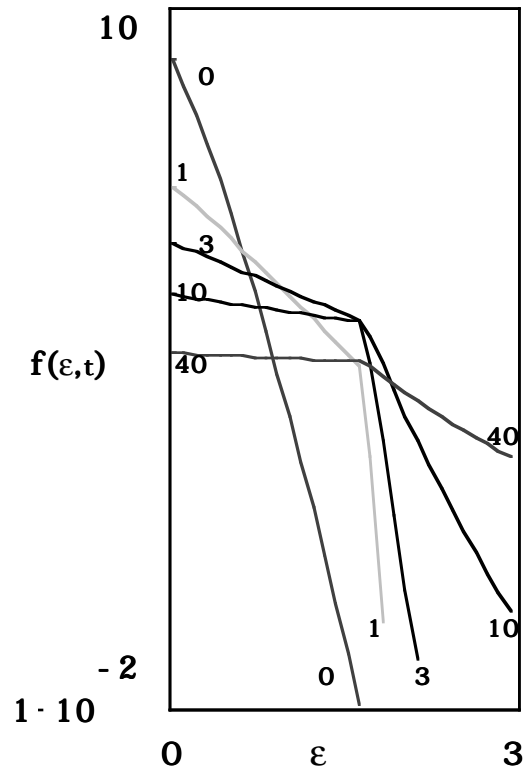
LOG[f(ε,t)] vs. ε (eV),

E/N = 0 to 100 Td
    in 20 μs,
labeled by time step,
Δt = 0.625 μs,
210 collisons/Δ t,
ω/ν < .009

Figure 2: 100 Td, 20 μs ramp
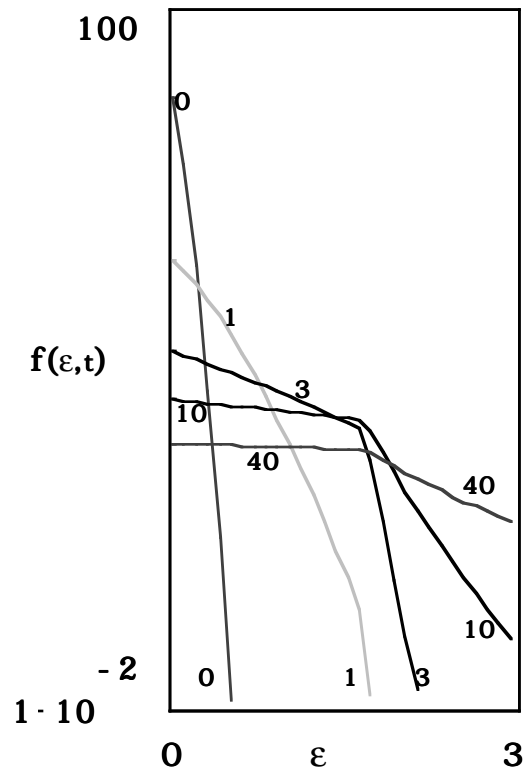
LOG[f(ε,t)] vs. ε (eV),

E/N = 0 to 100 Td
    in 2 μs,
labeled by time step,
Δt = 62.5 ns,
21 collisions/Δ t,
ω/ν < .09

Figure 3: 100 Td, 2 μs ramp

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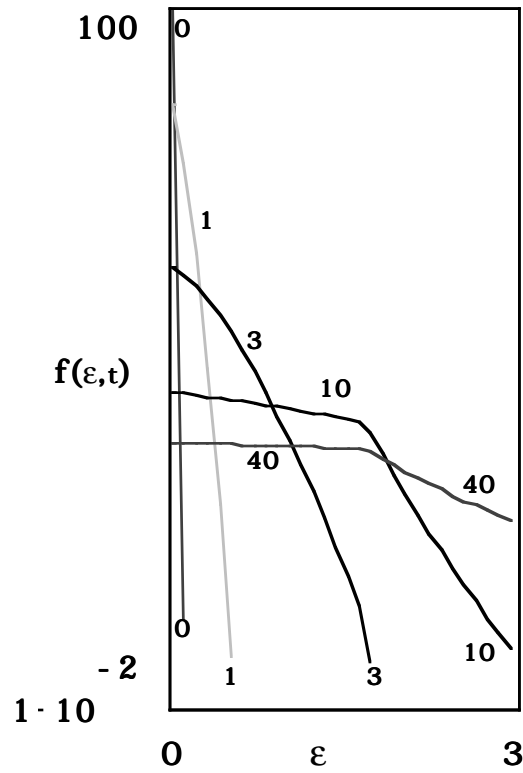
LOG[f(ε,t)] vs. ε (eV),

E/N = 0 to 100 Td
    in 0.2 μs,
labeled by time step,
Δt = 6.25 ns,
2 collisions/Δ t,
ω/ν < 0.9

Figure 4: 100 Td, .2 μs ramp
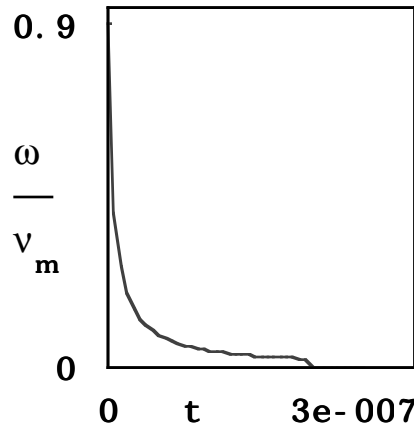
Figure 5: ω/ν_m for example of Figure 3

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$f(\varepsilon,t)$
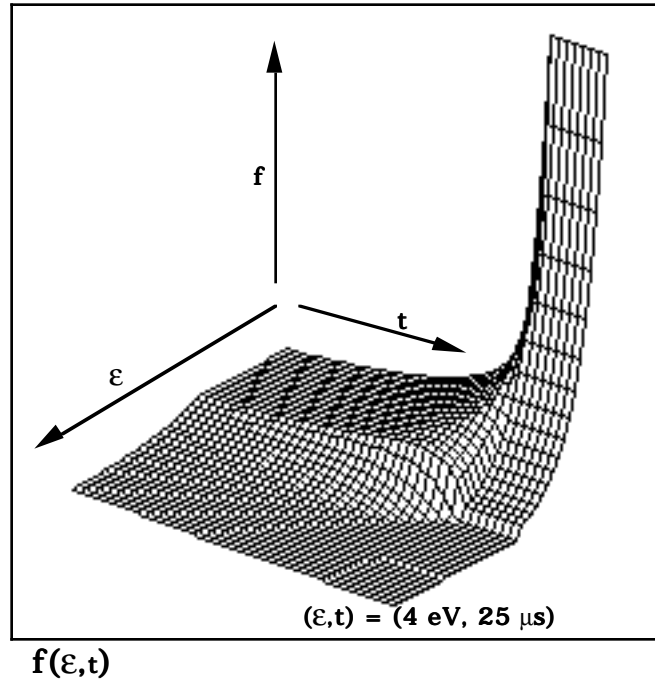
$(\varepsilon,t) = (4\ eV,\ 25\ \mu s)$

Figure 6: Field Collapse from 100 Td in 20 μs
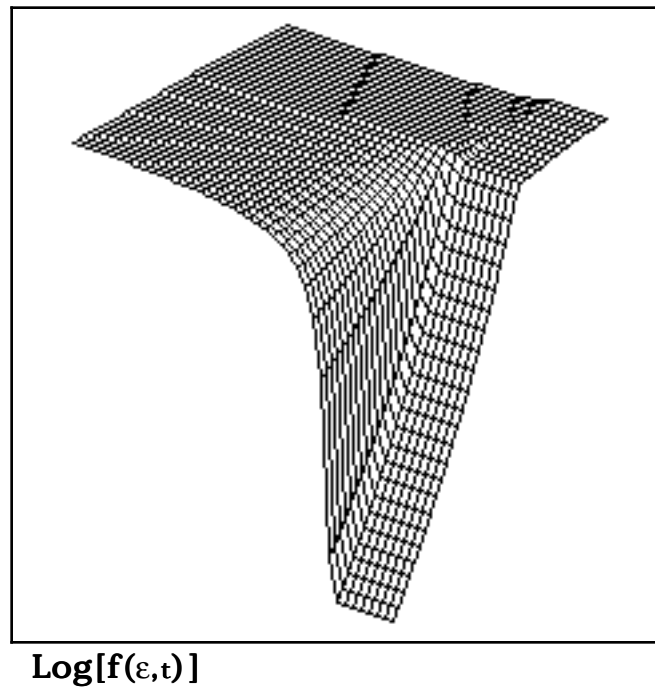
$Log[f(\varepsilon,t)]$

Figure 7: Log representation of Figure 6

16
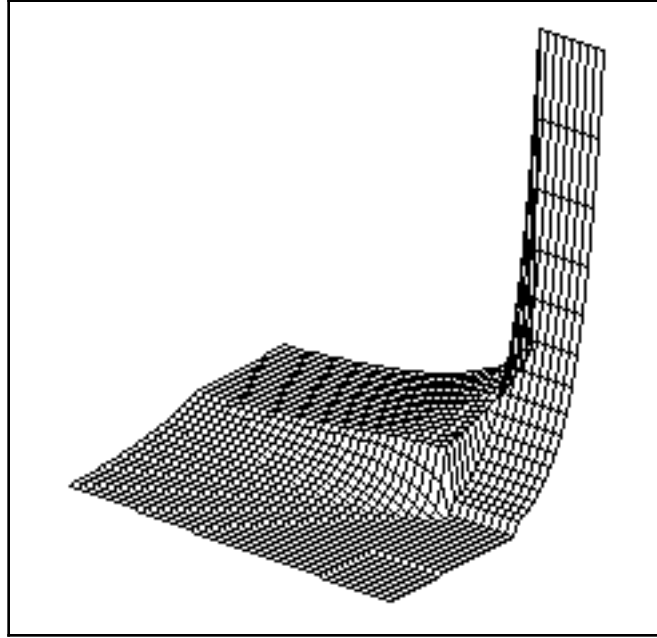
f(ε,t)

**Figure 8: Field Collapse from 100 Td in 2 μs
(ε,t) from (0,0) to (4 eV, 2.5 μs),**
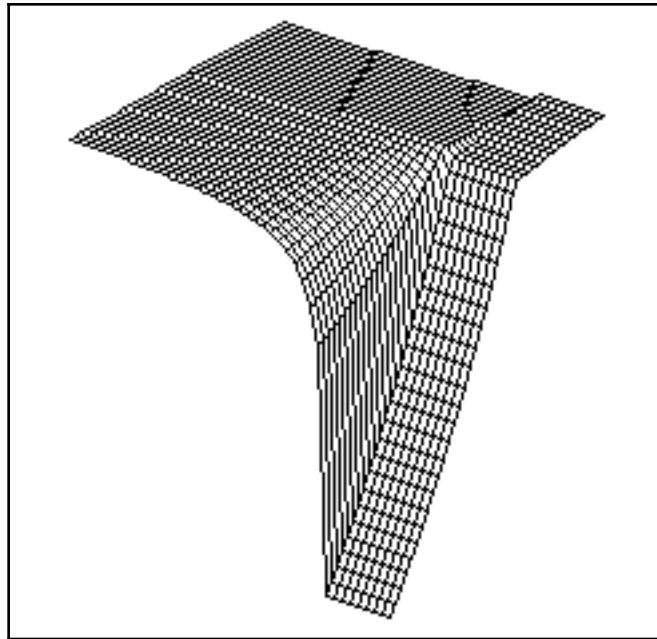
Log[f(ε,t)]

Figure 9: Log representation of Figure 8

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f(ε,t)
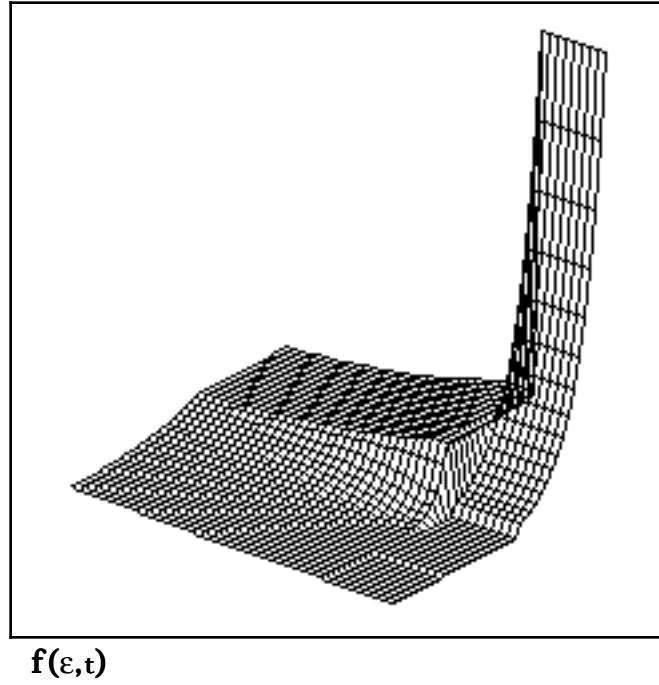
Figure 10:   Field Collapse from 100 Td in 0.2 μs
(ε,t) from (0,0) to (4 eV, 0.25 μs)

Log[f(ε,t)]
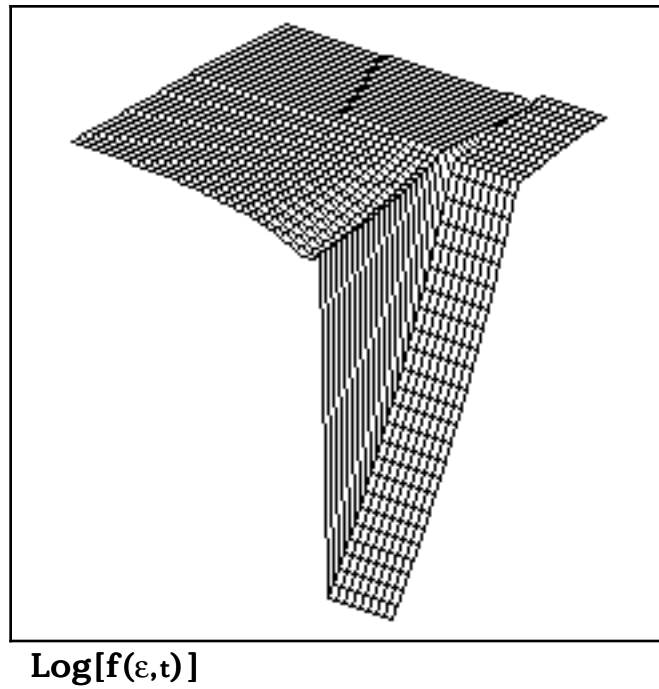
Figure 11:   Log representation of Figure 10
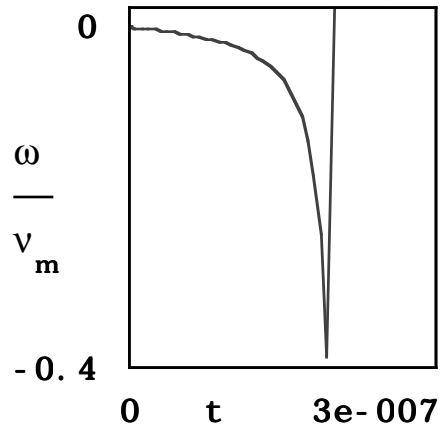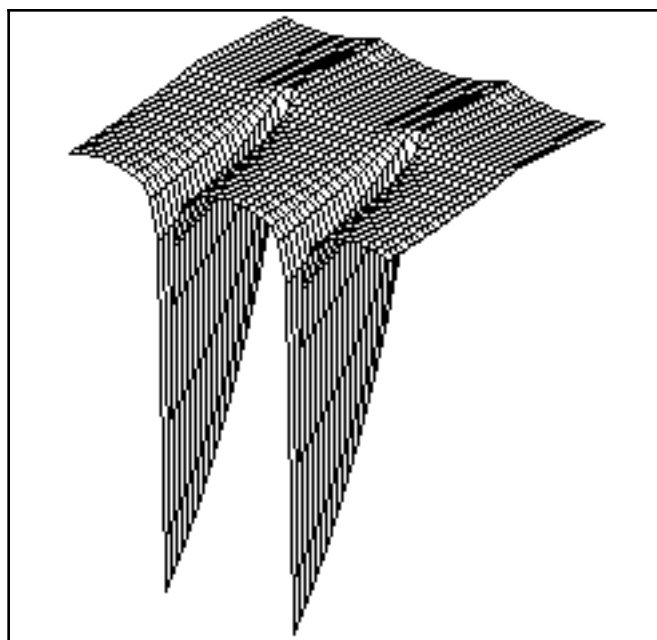
Figure 12: ω/ν_m **for example of Figure 10**

The last two examples are for E/N a sine wave with phase angle ϕ after t = 0. E/N prior to t = 0 is steady at 100 * sin(ϕ) Td. These examples have $\phi = \pi/4$. Figures 13 through 18 show these cases.
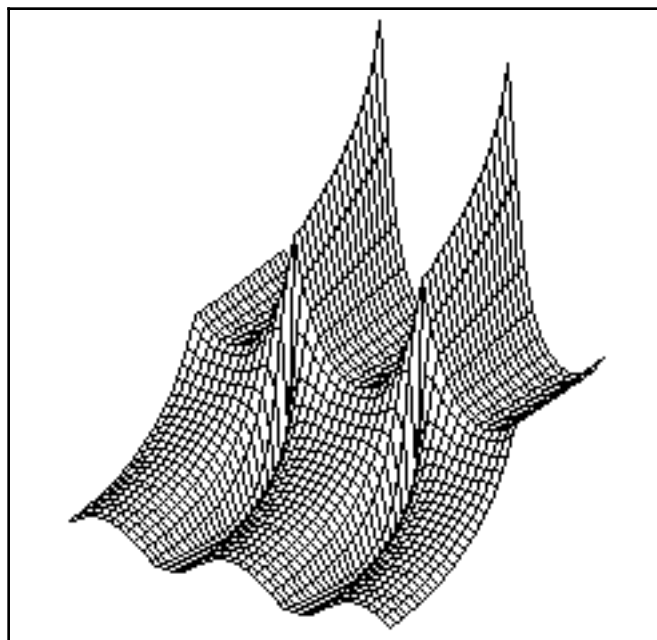
f(ε,t)

Figure 13: 100 Td, 50 kHz sine wave to (ε,t) = (4 eV, 25 μs)

Log[f(ε,t)]

Figure 14: Log representation of Figure 13

f(ε,t)
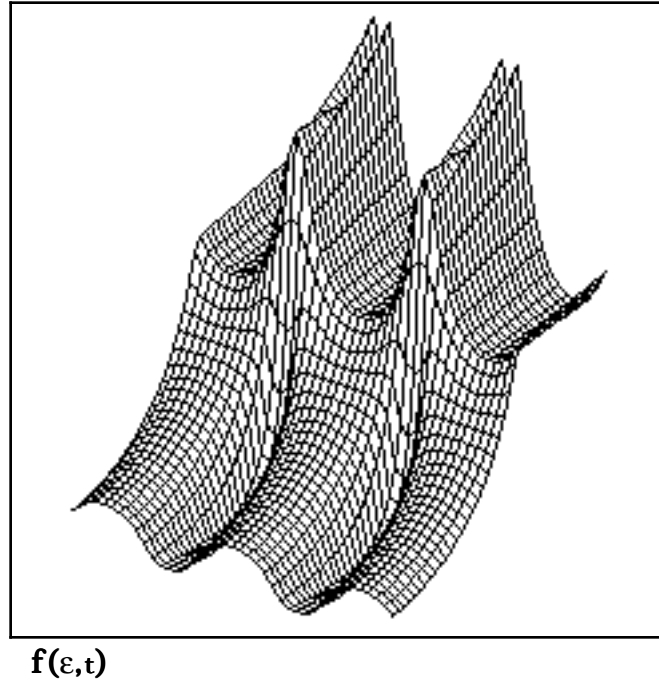
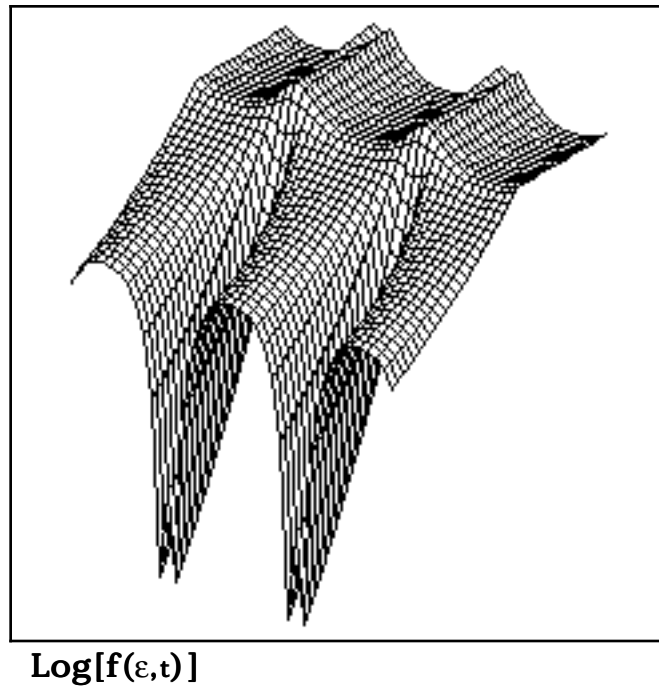**Figure 15: 100 Td, 500 kHz sine to (ε,t) = (4 eV, 2.5 μs)**

Log[f(ε,t)]

Figure 16: Log representation of Figure 15

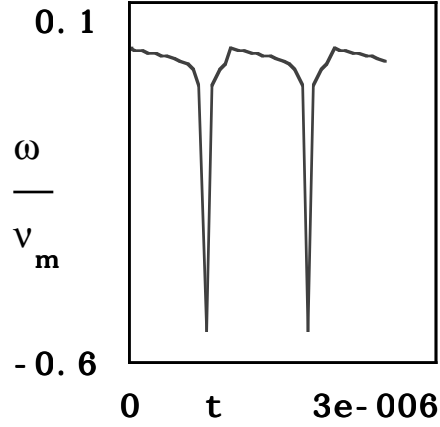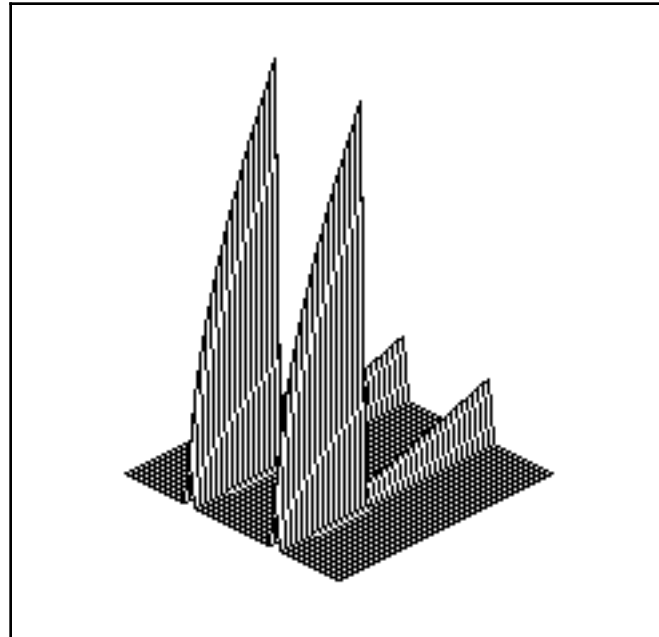**Figure 17:** ω/v_m for example of Figure 15

Ratio: $[B(\varepsilon, E(t)/N) @ \omega = 0] / B(\varepsilon, t)$

Figure 18: Ratio of $B(\varepsilon,t)$ with ω set to zero, to actual $B(\varepsilon,t)$ for example of Figure 15
[ratio = 1 at rectangular base, ridges along high ω/v_m]

    The microstructure developing along the node lines in the 500 kHz example is a mathematical artifact due to the comparable magnitudes of ω and v_m at those locations.

Conclusions

The function $B(\varepsilon,t)$ shown as equation (4) is presented as the logarithmic slope in energy of the electron distribution function in the case of a slightly ionized uniform gas with both inelastic collisions and time varying electric field. This function was derived from the Boltzmann equation through a sequence of approximations, and its validity relies on the condition $|\omega/\nu_m| < 1$.

In this analysis the frequency function $\omega(t)$, defined in equation (6) on the basis of the electric field as shown in equation (11), encapsulates the purely temporal effects of E(t) on the electron distribution. In addition the instantaneous magnitude of E/N also has an impact on the electron distribution. Figure 18 clearly shows how a time dependent solution can be quite different from a sequence of static solutions each at the instantaneous E(t)/N.

The model $B(\varepsilon,t)$ has physically reasonable limiting behavior, and the case of massless electrons in a static field was favorably compared to published calculations and data for a variety of gases in a previous work.[7] Any approximate analysis such as this one strives to find an appropriate balance between a convenient and widely applicable result on the one hand, and accuracy on the other. Quantifying the degree to which this model approaches that balance is left to future work.

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6)   I am grateful to Dr. Britton Chang of LLNL for his suggestions on this point.

7)   M. Garcia, B. Chang, "Analytical Boltzmann Moments for Electrons in N_2-O_2-H_2O Gas Mixtures," LLNL UCRL-ID-117124, presented at the IEEE International Conference on Plasma Science at Santa Fe, NM on 6 - 8 June 1994.